

TROPOSPHERIC OZONE FROM TOMS: PROVIDING THE FIRST DEPICTIONS OF THE EXTENT OF GLOBAL POLLUTION

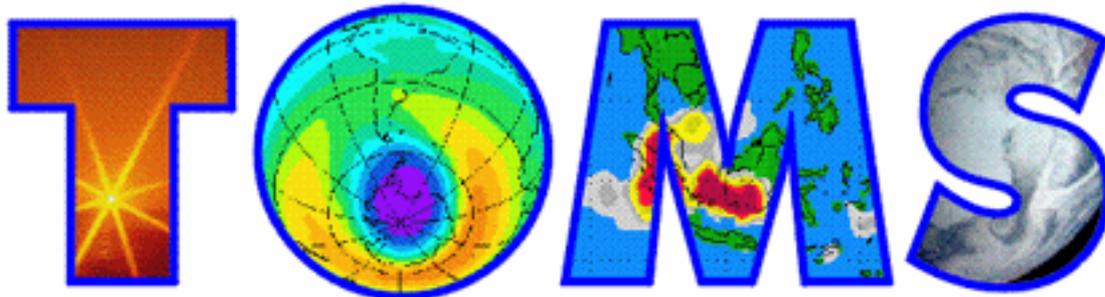
Jack Fishman¹, Sushil Chandra², Anne M. Thompson², Jerry R. Ziemke²
Robert D. Hudson³, Michael J. Newchurch⁴
John K. Creilson¹, Amy E. Wozniak^{1,2}

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AGU Fall Meeting
San Francisco, CA
December 8, 2003

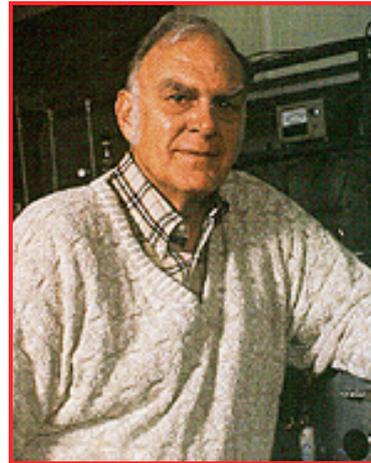
The Origin of Using Satellite Data to Study Tropospheric Ozone Can be Linked to Nobel-Prize Winning Research

from Nobel Prize press release:

The Royal Swedish Academy of Sciences has decided to award the 1995 Nobel Prize in Chemistry to

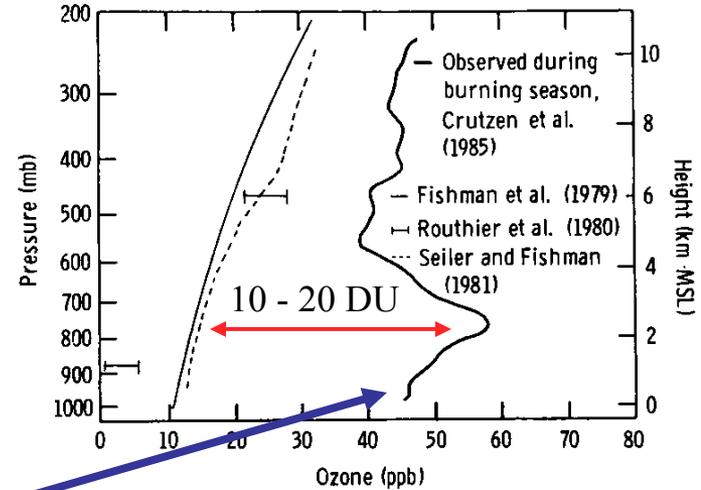
Paul Crutzen, Mario Molina and **F. Sherwood Rowland**

for their work in atmospheric chemistry, particularly concerning **the formation** and decomposition **of ozone**.



In his search for understanding the sources of ozone in the troposphere, Crutzen made the first comprehensive measurements trace gases where tropical biomass burning was occurring and found considerably higher concentrations than what had been published previously

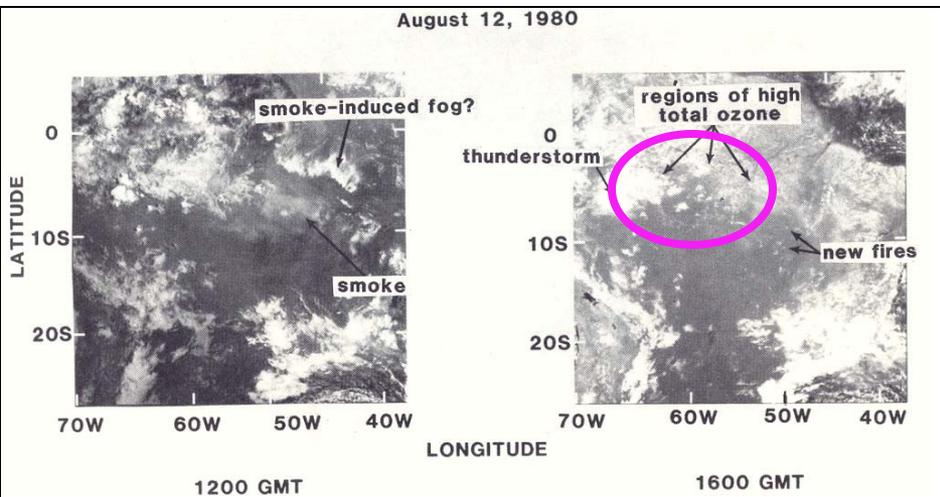
OZONE DISTRIBUTION AT SOUTHERN TROPICAL LATITUDES



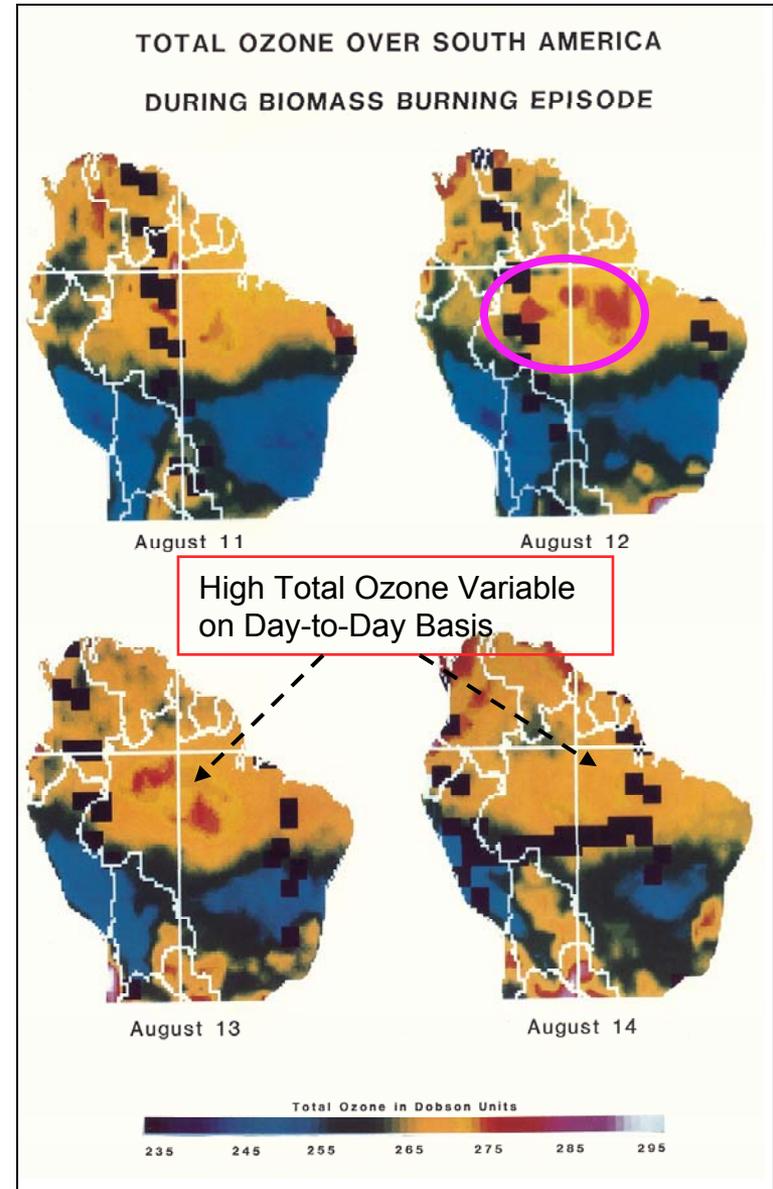
(from Fishman, Minnis & Reichle, *JGR*, 91, 1986)

- **Can the 10-20 Dobson Unit Enhancement Be Identified from TOMS Total Ozone Measurements?**
- Such Enhancements are Better Observed at Low Latitudes Due to Less Stratospheric Variability
- TOMS Precision is 1% (~ 3 DU)

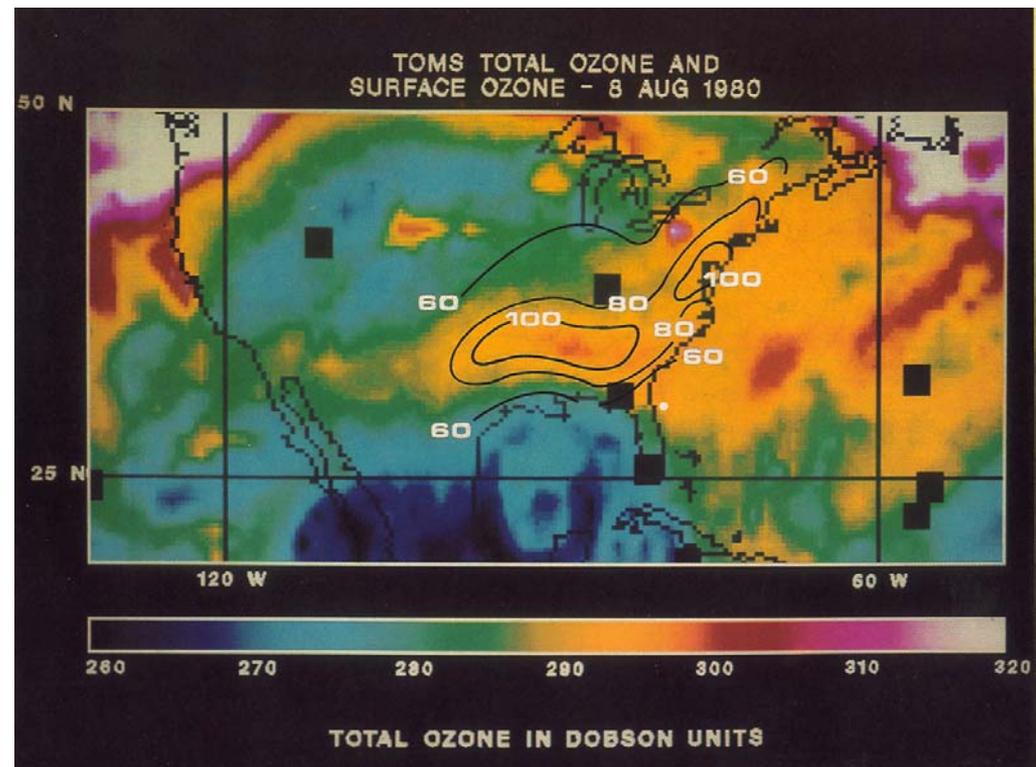
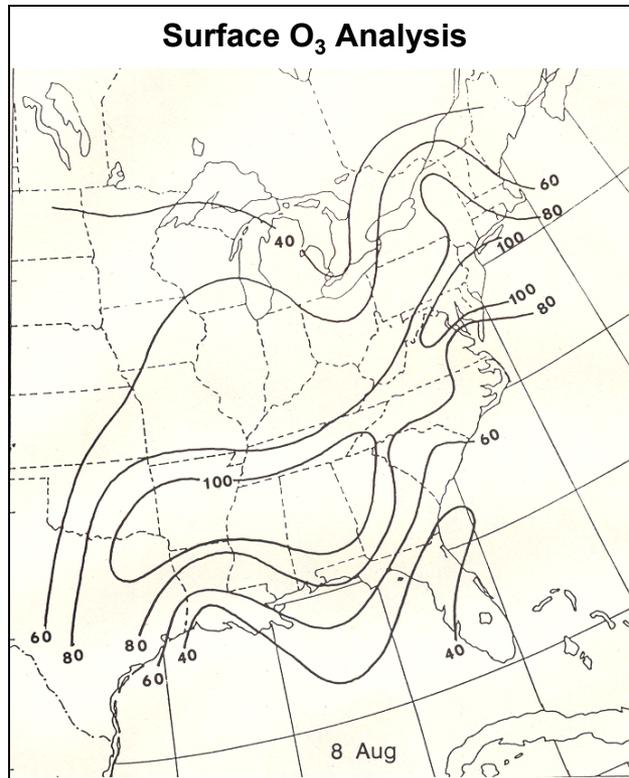
Enhanced Total Ozone Observed in Conjunction with Biomass Burning in 1980 Episode



(from Fishman, Minnis & Reichle, *JGR*, 91, 1986)

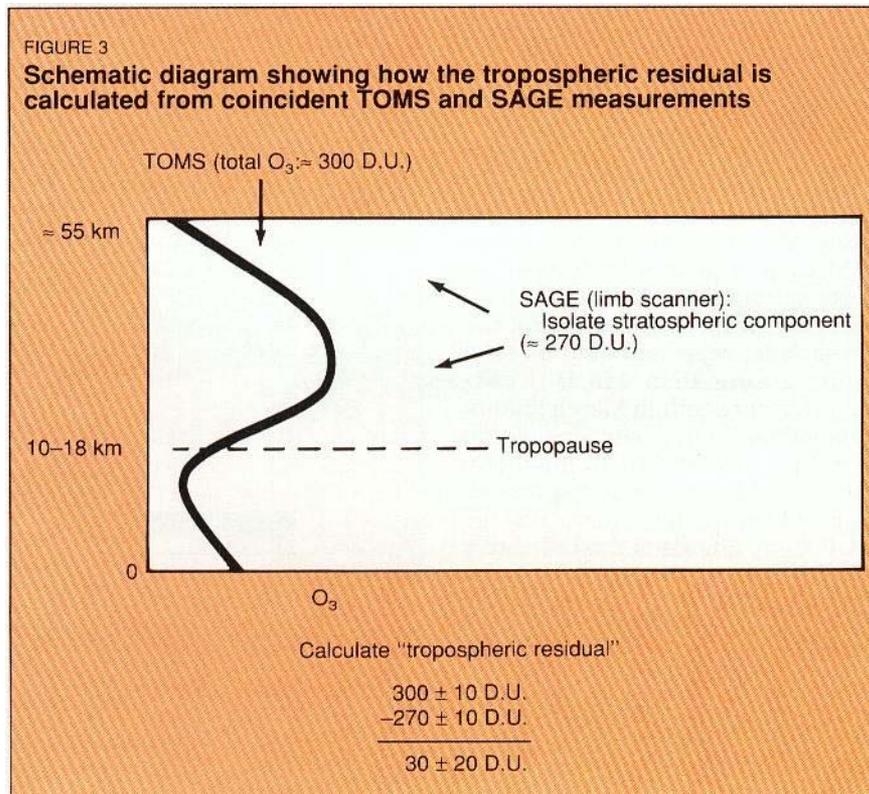


High Surface Ozone Concentrations During Pollution Episode Also Observed in TOMS Total Ozone



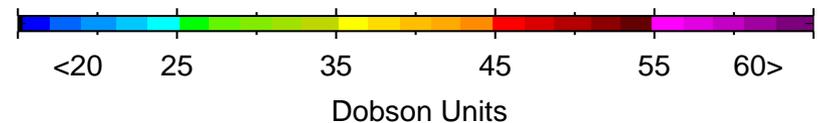
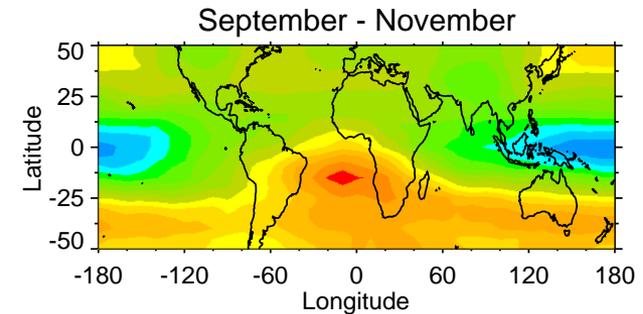
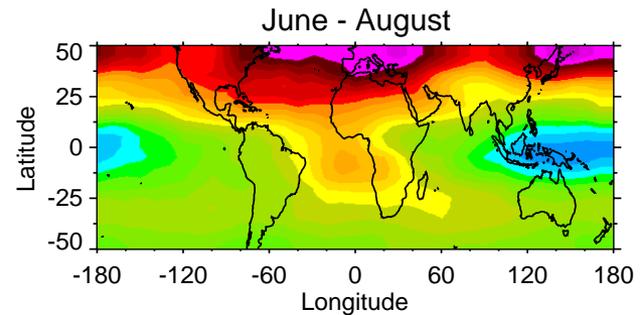
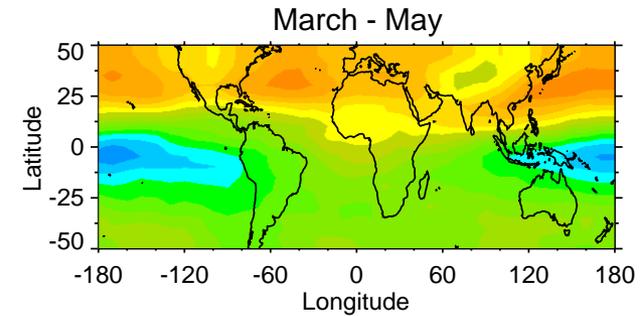
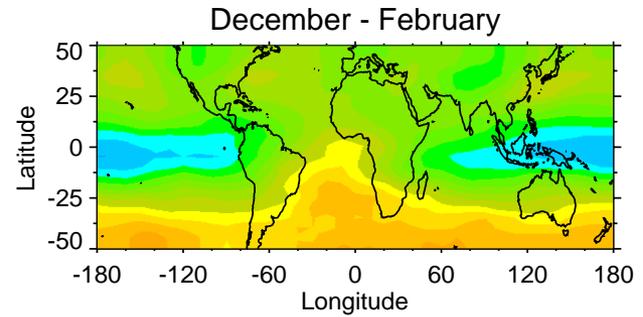
(from Fishman et al., *J. Clim. Appl. Met.*, **26**, 1987)

Separate Stratosphere from Troposphere to Compute Tropospheric Ozone Residual (TOR)



First Separation of TOMS Total Ozone to Derive Tropospheric Ozone Residual Used SAGE Measurements to Determine Stratospheric Ozone:

- Seasonal Climatologies Produced
 - Highest TOR in NH Summer
 - Tropical Enhancement in Austral Spring
- Data Too Sparse to Examine Interannual Variability



Other Techniques Use TOMS to Measure Tropospheric Ozone

- **Thompson and Hudson:**

Isolate “tropical” air in region of wave-one and then separate strat. & tropo. ozone using sonde data

- **Chandra and Ziemke**

TOR calculations using MLS and HALOE for stratosphere

Convective Cloud Differential (CCD)/Cloud Slicing

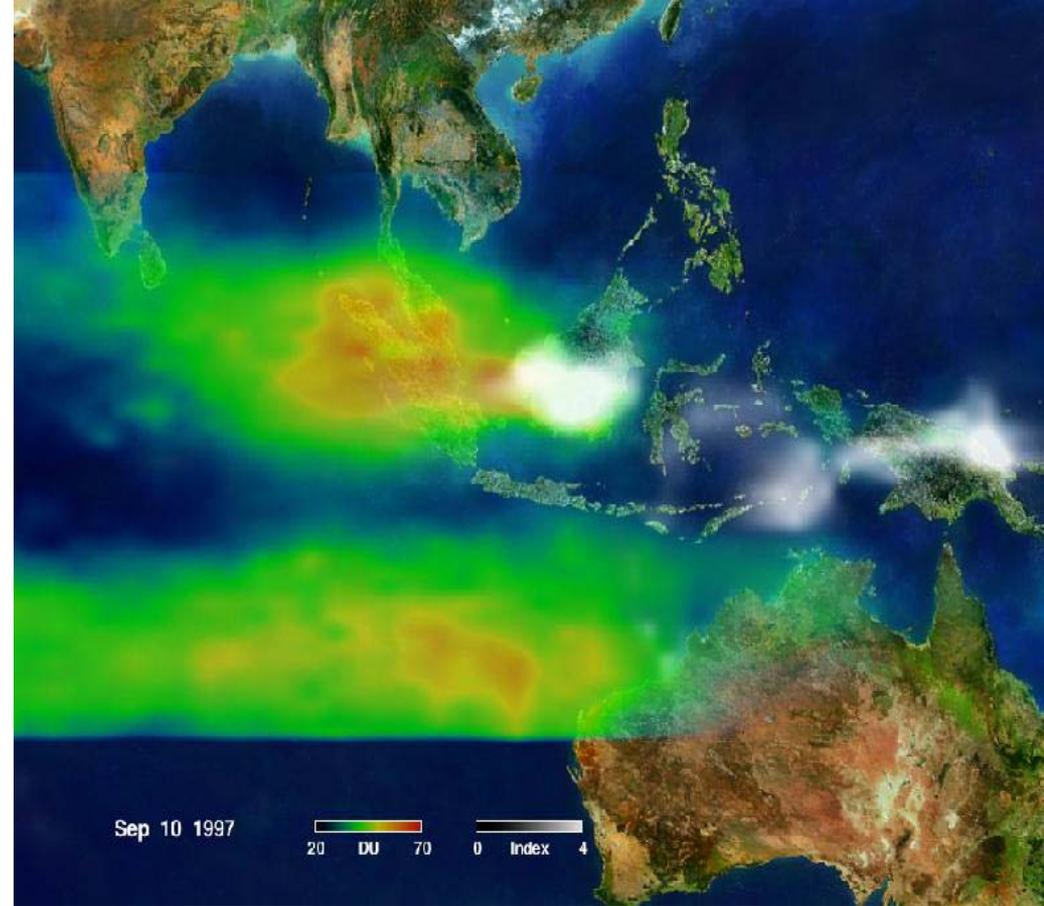
- **Newchurch and co-workers**

Scan-angle dependence method

Terrain-dependence to determine lower tropospheric ozone

Daily Maps: Modified-residual Method, Thompson & Hudson, 1999

- Isolate “tropical” air in region of wave-one; separate strat. & tropo. ozone using sonde data
- TOMS image, 10 Sept. 1997, resolves smoke from fires (gray), ozone (colors) during ENSO-related Indonesian fires



- **Scientific results:** (1) smoke, ozone transport traveling in different layers; (2) upper plume – Indonesian origin; lower plume – African; (3) two plumes merged, 22 Oct. 1997 [Thompson et al., *Science*, 291, 2128, 2001]

Ziemke and Chandra Developed TOR and CCD Methods

- 1. Tropospheric Ozone Residual (TOR) Method

$$\text{TOR} = \text{Total Column Ozone} - \text{Stratospheric Column Ozone}$$

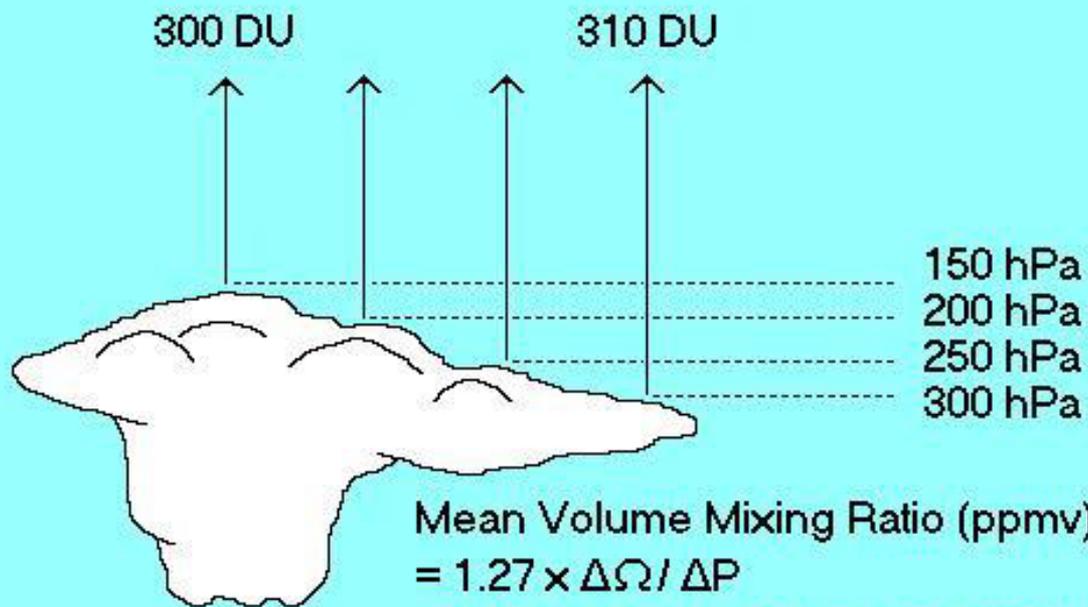
- Total column ozone is measured by the TOMS instrument.
- Stratospheric column ozone is measured by SAGE, **HALOE** or **MLS**.

- 2. Cloud Slicing/CCD Method

- UV radiation measured by TOMS is opaque to the dense water vapor clouds in the troposphere. This allows measurement of stratospheric column from high reflecting clouds near the tropopause and total column ozone from nearby high reflecting scenes. Again,

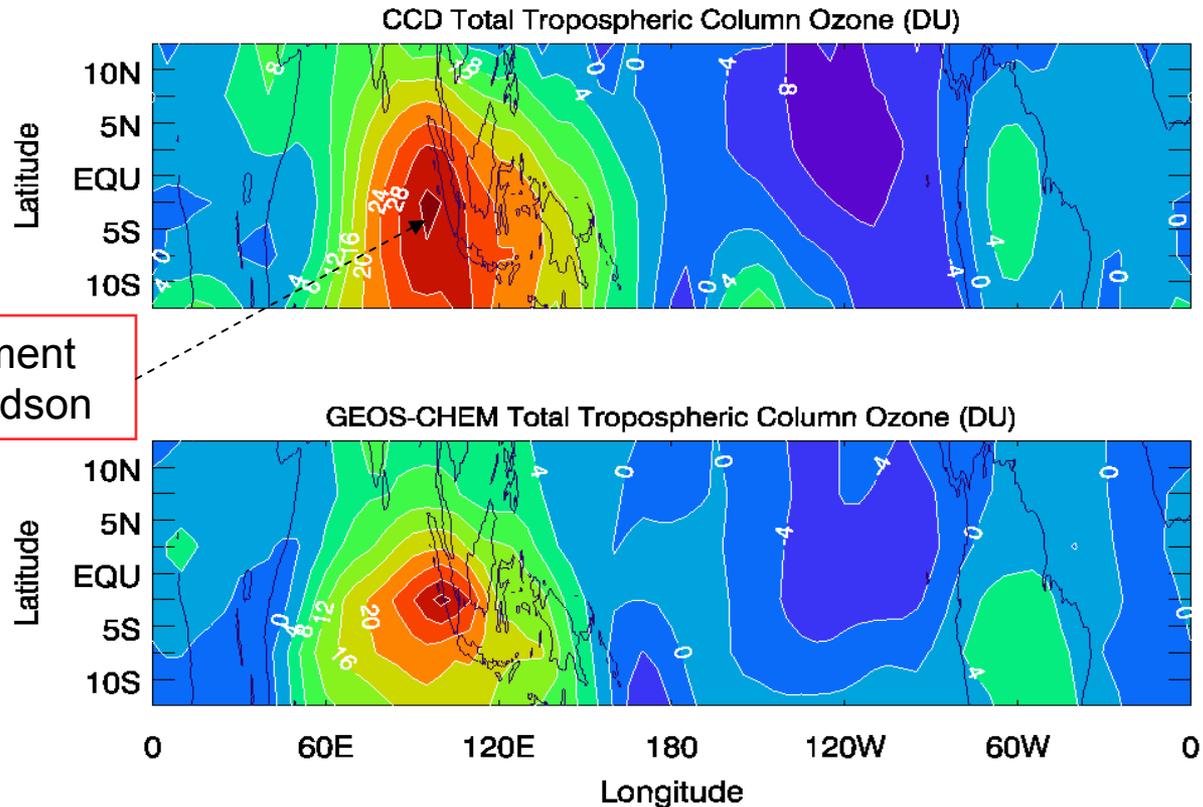
$$\text{TOR} = \text{Total Column Ozone} - \text{Stratospheric Column Ozone}$$

Using Cloud Tops To Obtain Mean O3 Volume Mixing Ratio



$$\begin{aligned} \text{Mean Volume Mixing Ratio (ppmv)} &= 1.27 \times \Delta\Omega / \Delta P \\ &= 1.27 \times 10 \text{ DU} / 150 \text{ hPa} = 0.085 \text{ ppmv} \end{aligned}$$

El Nino Related Changes in Tropospheric Ozone (Oct97 Minus Oct96)

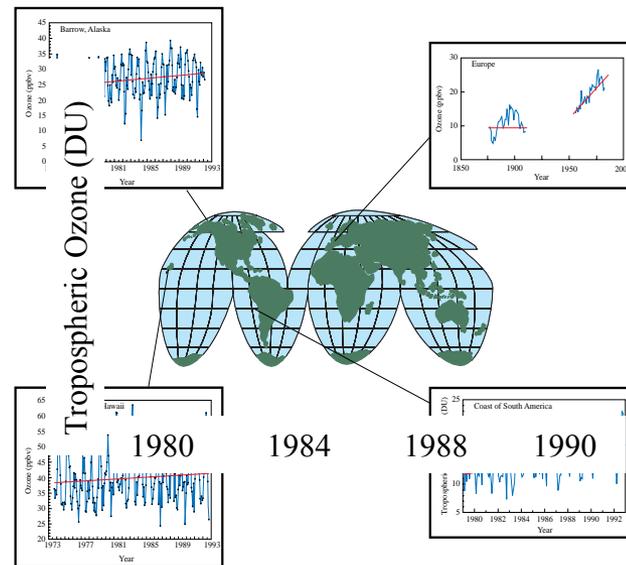


Shown is the effect of El Niño on tropical TOR. Top: TOMS CCD TOR change from October 1996 to October 1997 (i.e., shown is October 1997 minus October 1996 TOR). Bottom: Same as top frame but instead based on GEOS-CHEM model TOR. The high degree of consistency between the two suggests that the GEOS-CHEM model provides a useful tool to separate the influence of dynamics and biomass burning on TOR. The model suggests that about half of El Niño-induced ozone arises from dynamics and half from biomass burning. (For more details, please see the paper *Chandra et al.*, JGR, July, 2002.)

Various Techniques Have Used Terrain Information to Derive Tropospheric Information:

Confirmation that TOMS Sees into the Lower Troposphere

- Jiang and Yung (1996) Used Data Adjacent to Andes to Produce Tropospheric Ozone Trend



- Newchurch and Co-workers Examined Many Locations to Examine Seasonality
- Fishman et al. (2003) Used Northern Africa Measurements to Infer Profile for Validation

Currently Published in *Atmospheric Chemistry and Physics*:

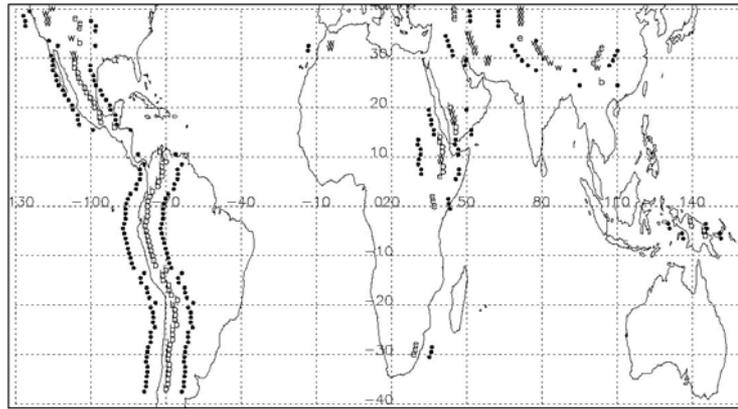
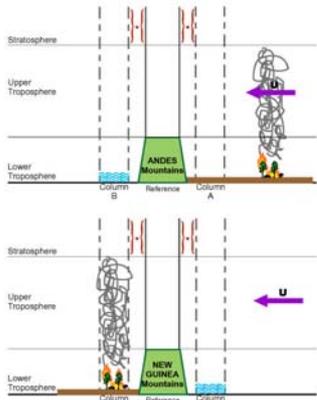
Problems regarding the Tropospheric O₃ Residual method and its interpretation in Fishman et al. [2003]

A.T.J. de Laat and I. Aben

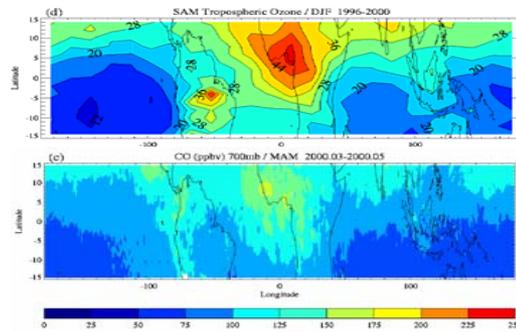
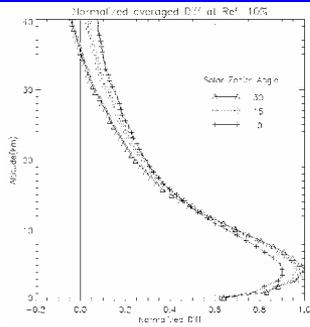
“We will show that it is possible to obtain a tropospheric O₃ column that is very similar to what is being presented in Fishman et al. [2003], solely based on the Logan [1999] tropospheric O₃ climatology and an estimate for the tropopause heights without using satellite data.”

The Above Statement is grossly erroneous and can be seen through a simple visual comparison of the data sets!

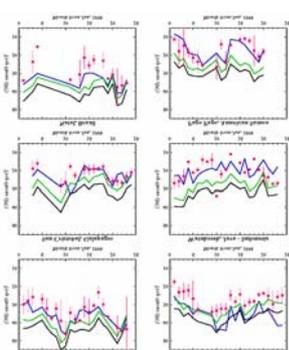
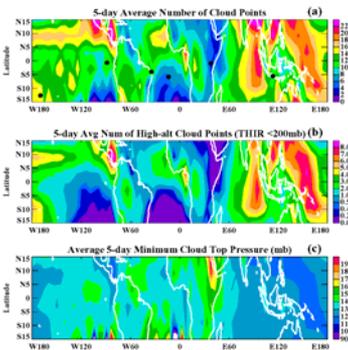
Newchurch and co-workers: Terrain-height differences, Scan Angle Method (SAM), and Clear Cloudy Pairs (CCP)



Lower Tropospheric ozone near Mountain ranges.
 Good agreement with sondes and fire counts.
 Seasonal variation computed.
 [Kim and Newchurch, GRL, 1996; Kim and Newchurch, JGR, 1998; Newchurch et al., JGR, 2001]



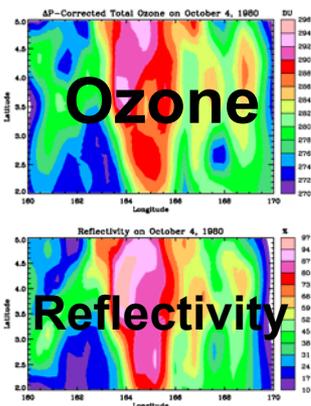
Scan Angle kernel peak at ~5km.
 Good agreement w/ sondes, MOPPITT CO, ATSR fire counts, GEOS-Chem model.
 Resolves N. Atlantic paradox.
 Monthly tropical, lower-tropospheric ozone columns.
 [Kim et al, JAS, 2001; Kim et al., JGR, submitted, 2003]



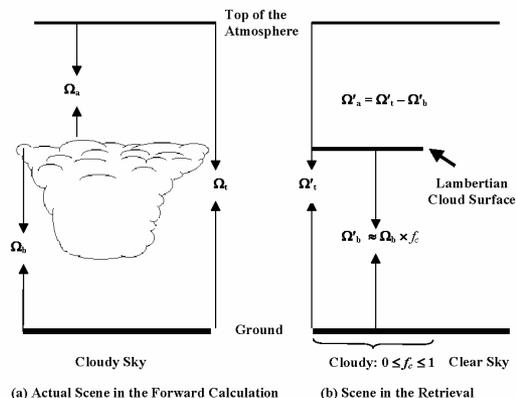
CCP: Use all high-alt clouds (low THIR temp or high TOMS 380 nm reflectivity); stratosphere constrained by all clouds, not necessarily zonal flat. Good agreement with sondes (3 ± 1 DU).
 [Newchurch et al., ACP, 2003.]

All data and references are available on nsstc.uah.edu/atmchem

Newchurch et al.: Cloud anomalies, retrieval accuracy, and comparison of all tropospheric methods



Cloud anomalies (ozone/reflectivity correlation, positive and negative) in TOMS: Frequency of occurrence, morphology, explanation. [Liu, UAH Dissertation, 2003; Liu et al., ACP, 2003]



Retrieval accuracy. Explanation of retrieval errors above clouds. [Newchurch et al., JGR, 2001a; Newchurch et al., JGR, 2001b; Liu et al., JQSRT, 2003]

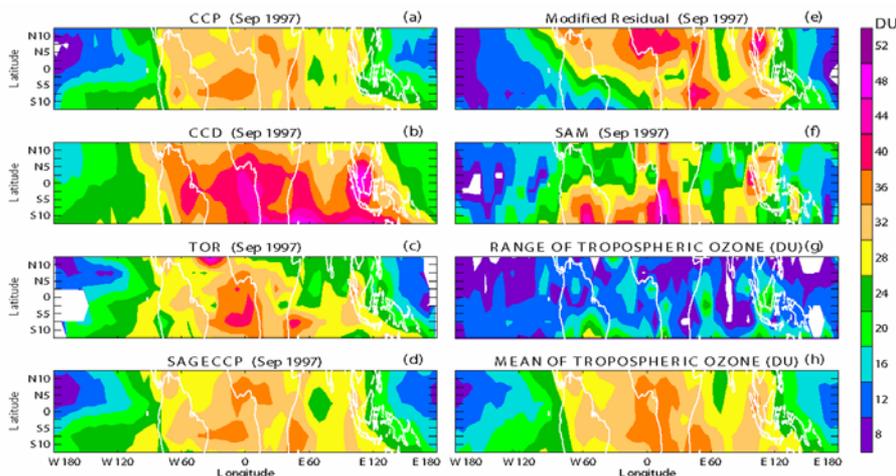


Figure 1 Tropical tropospheric ozone derived by six different methods along with the range and mean in September 1997. CCP results from Clear-cloudy Pairs of observations, CCD results from our calculation of the Convective Cloud Differential method prescribed by [Ziemke et al., 1998], 5DU subtraction is applied as suggested by the author. TOR results from our calculation of the Tropospheric Ozone Residual (TOMS-SAGE) prescribed by [Fishman and Larsen, 1987]. SAGE-CCP results from a hybrid of the CCP method where high clouds are present and SAGE stratospheric ozone where clouds are absent. MR method are described in [Thompson and Hudson, 1999], SAM results from differences of TOMS clear-sky total ozone columns taken at high scan angles and nadir scan angles as prescribed by [Kim et al., 2000].

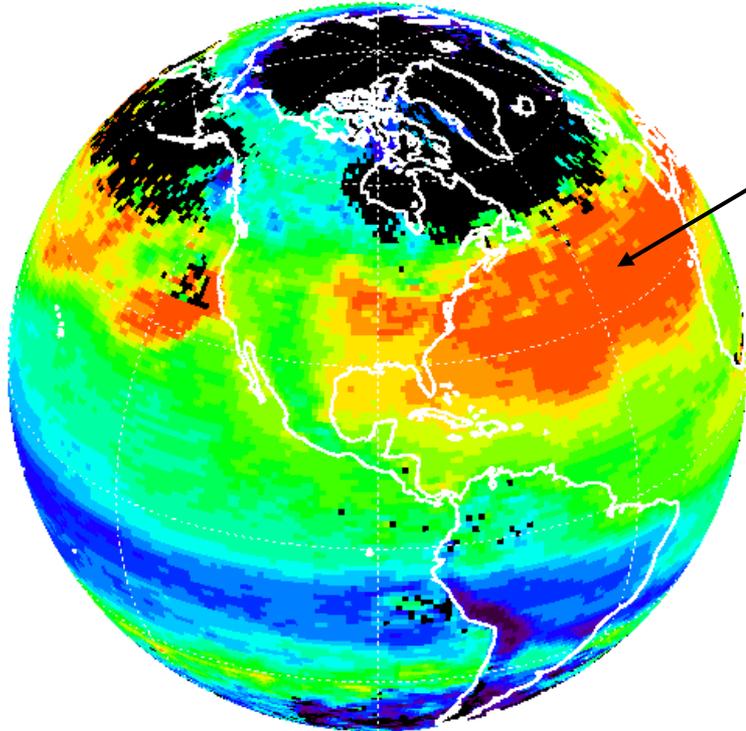
Comparison of 6 methods: CCP, CCD, TOR, SAGECCP, Modified Residual, SAM, sondes, GOME, lidar, model. Statistics: Monthly RMS diff 4 – 12 DU Pacific, 6 – 18 DU Atlantic. Each method has strengths/weaknesses. No single method best over all times/locations w.r.t. sondes.

[Sun, UAH Dissertation, 2003]

All data and references are available on nsstc.uah.edu/atmchem

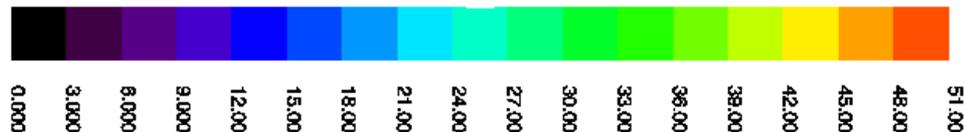
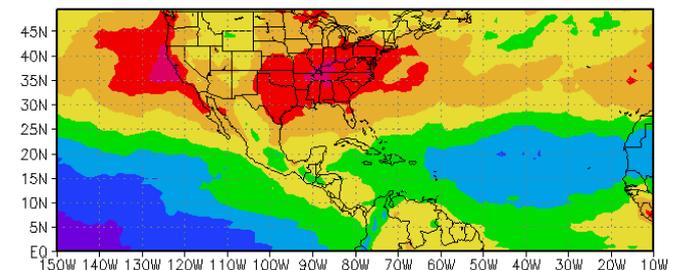
Tropospheric Ozone From TOMS

TOMS/MLS Tropospheric Column O₃ (DU) July 1992



Tropospheric column ozone from TOMS and MLS which shows large enhancement over eastern U.S. and North Atlantic in July 1992

Tropospheric Ozone Residual from TOMS/SBUV for July 1992



Improvement of Spatial Resolution Using TOMS/SBUV Residual

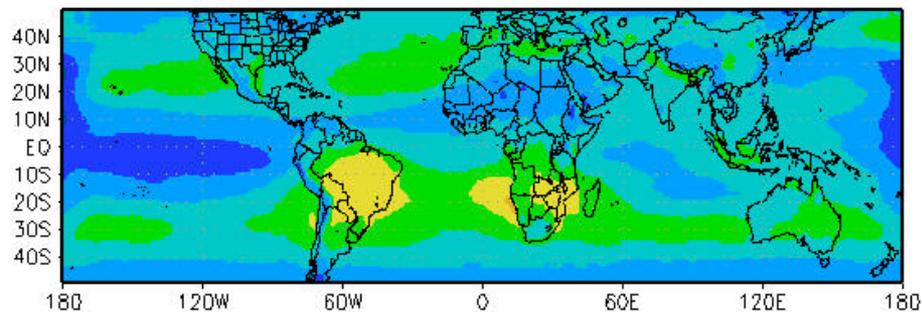
- Apply “Empirical Correction” to SBUV Profile
- Enough Information to Examine Interannual Variability

Other Satellite Data Sets Are Required To Separate Tropospheric Ozone from Total Ozone Measurements

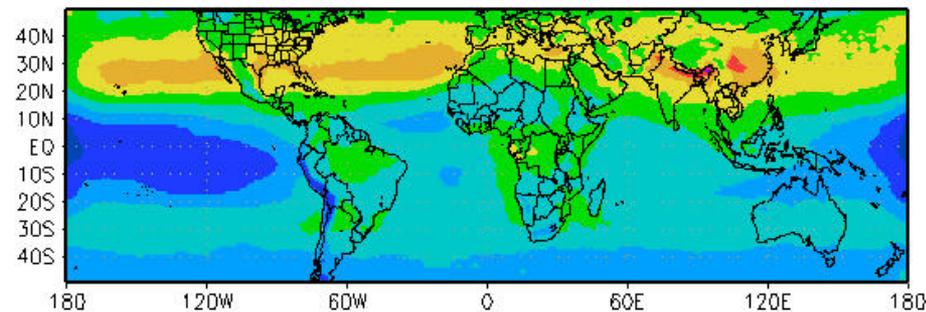
- SAGE: Good Vertical Resolution; Poor Spatial Coverage
 - HALOE: Good Vertical Resolution; Poor Spatial Coverage
 - MLS: Vertical Resolution Only >68 mb; Relatively Good Spatial Coverage
Only One Archived Layer below 100 mb
 - SBUV: Poor Vertical Resolution; Good Spatial Coverage
Archived Layers: 1000–253 mb; 253–126 mb; 126–63 mb
Stratospheric Fields Generated from 5 Days of Data
-
- **SAGE/TOMS TOR:** ~ 30,000 Coincident Observations 1979–1991 [Fishman & Brackett, 1997]
~ **10 data points per 5° x 10° grid box** for seasonal climatology
 - **SAGE/SBUV TOR:** Use Every TOMS Observation (up to 28,800 per day)
~ **1500 data points per 1° x 1.25° grid box** for seasonal climatology

Seasonal Depictions of Climatological Tropospheric Ozone Residual (TOR) 1979-2000

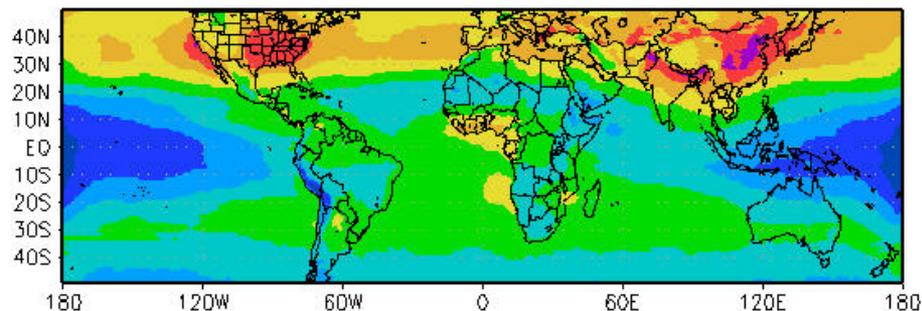
December - February



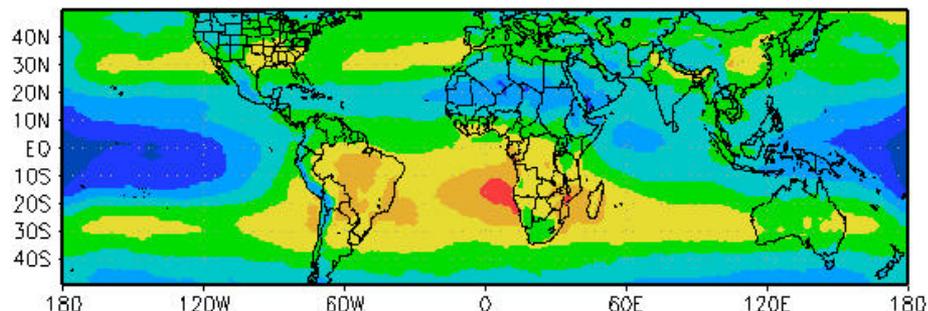
March - May



June - August



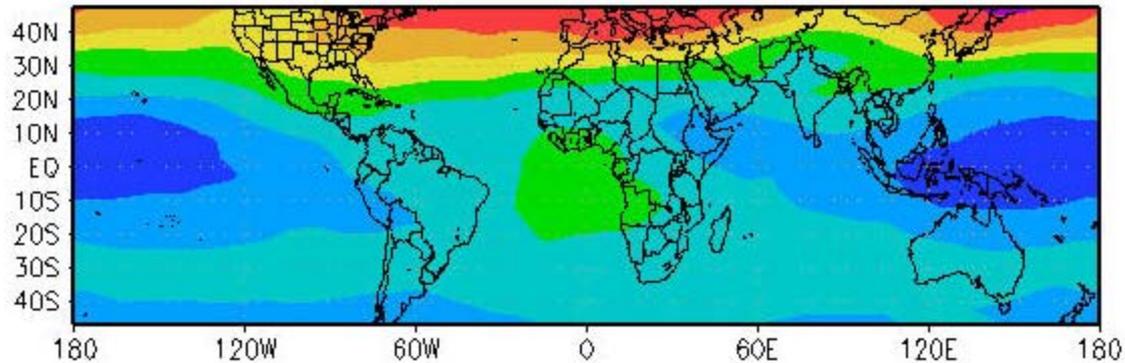
September - November



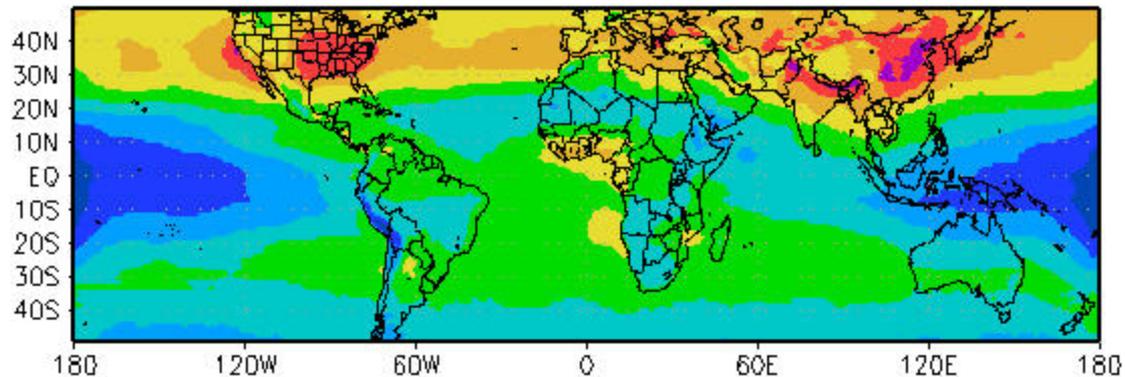
Dobson Units (DU)

Comparison of TOMS/SAGE TOR with TOMS/SBUV TOR: Regional Enhancements Not Previously Seen Now Found

TOMS/SAGE TOR: June-July-August Climatology (1979-1991)

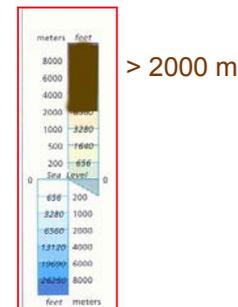
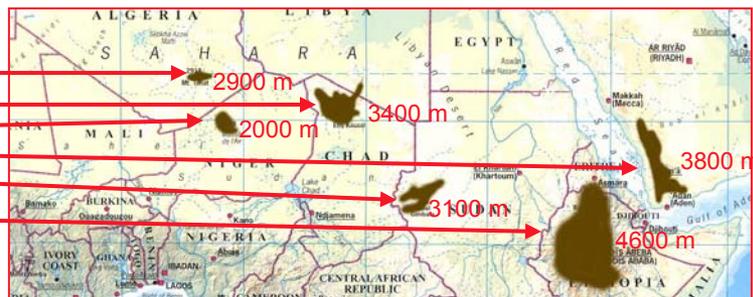
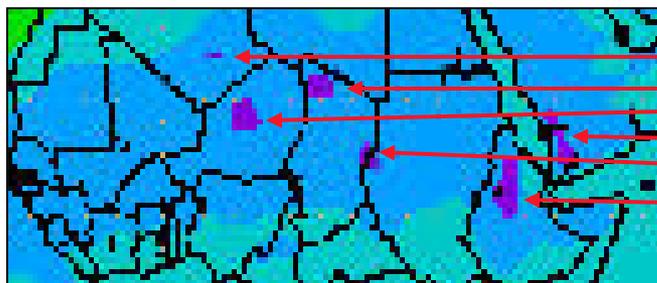


TOMS/SBUV TOR: June-July-August Climatology (1979-1991)

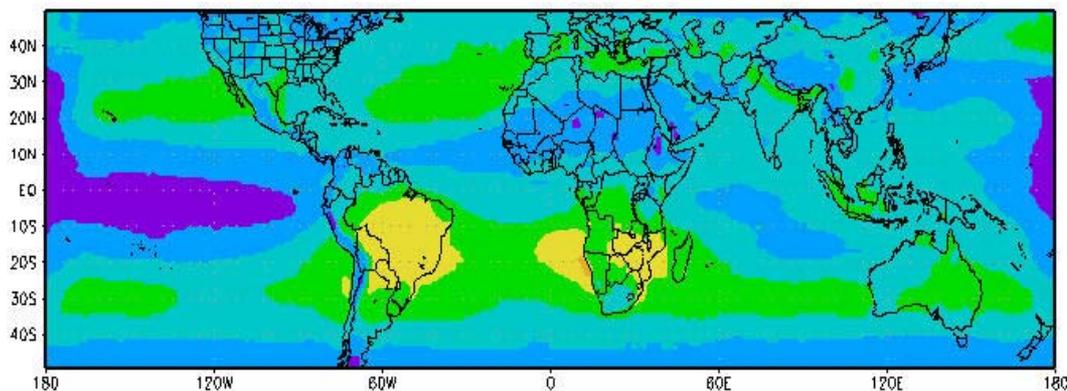


Dobson Units (DU)

Lower TOR over North African Desert Regions Coincident with Higher Elevations



December-February TOR

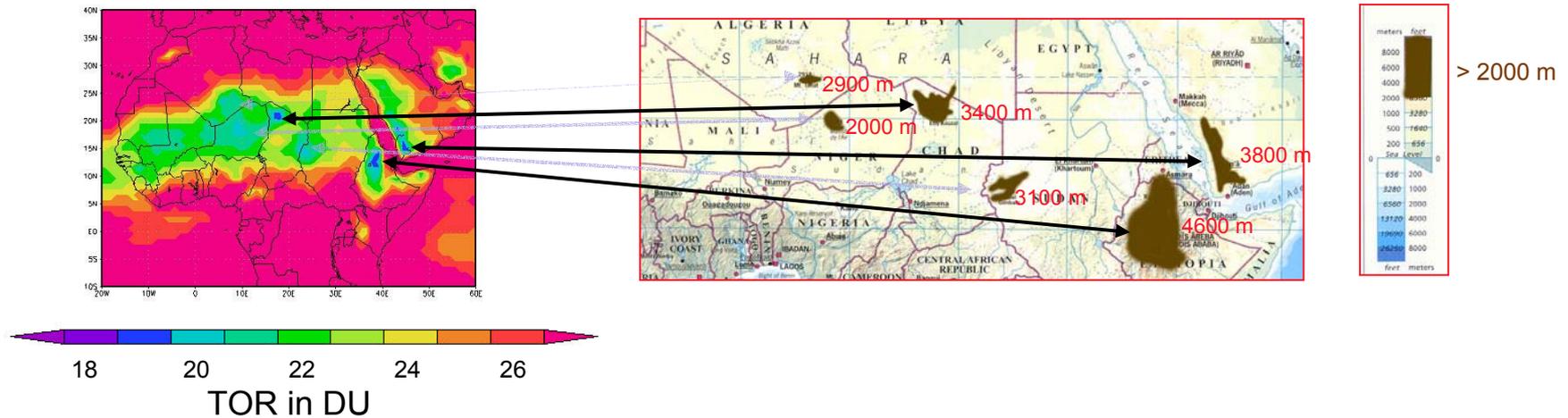


TOR in DU

Implications:

- TOMS Capable of Isolating Small (Regional) Scale Features
- ~ 3 DU for $\int^{2\text{km}} dz \Rightarrow \sim 20$ ppb in pbl
- Information can be used to validate O_3 backscatter sensitivity in boundary layer over cloudless unpolluted area

Higher Elevation Differences (3-4 km) Coincident with Greater O₃ Deficits (5-7 DU)

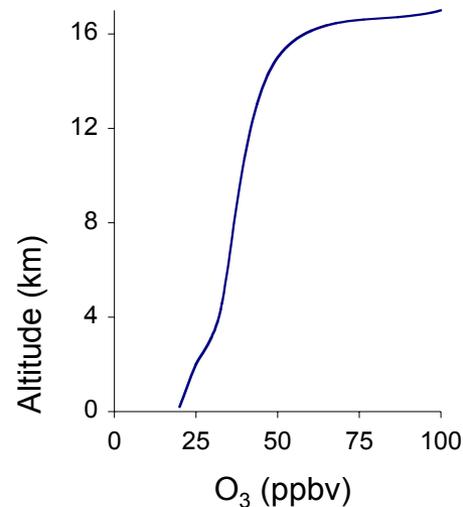


• Inferred Ozone Profile over North Africa Desert Region:

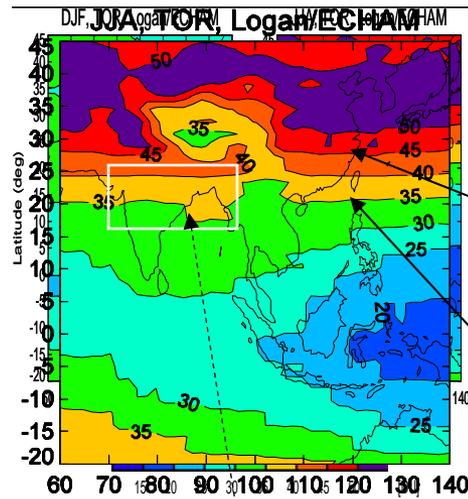
$$\int^{2 \text{ km}} [\text{O}_3] dz = \sim 3 \text{ DU}$$

$$\int^{4 \text{ km}} [\text{O}_3] dz = \sim 6 \text{ DU}$$

$$\int^{\text{Trop. } (\sim 17 \text{ km})} [\text{O}_3] dz = \sim 25 \text{ DU}$$

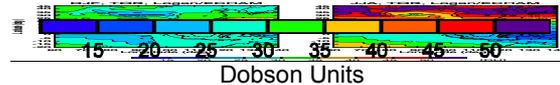


SBUV/TOMS TOR Provides Much Greater Spatial Resolution than Climatology Combined with Model Tropopause Heights

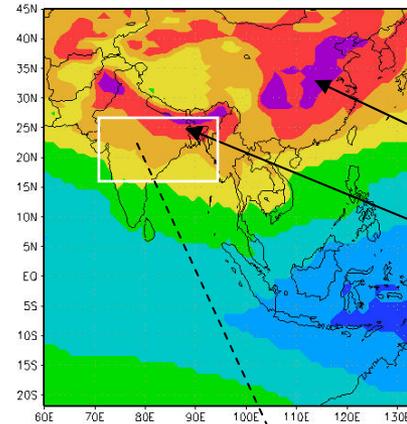


Calculated distribution mirrors zonal nature of input climatology

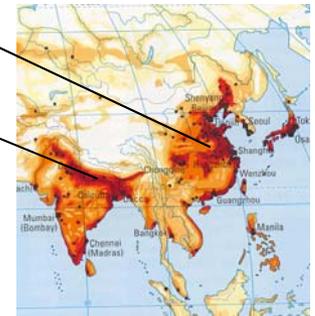
Data points going into white box



JJA TOR (Fishman et al., 2003)



Satellite data reflect population distribution



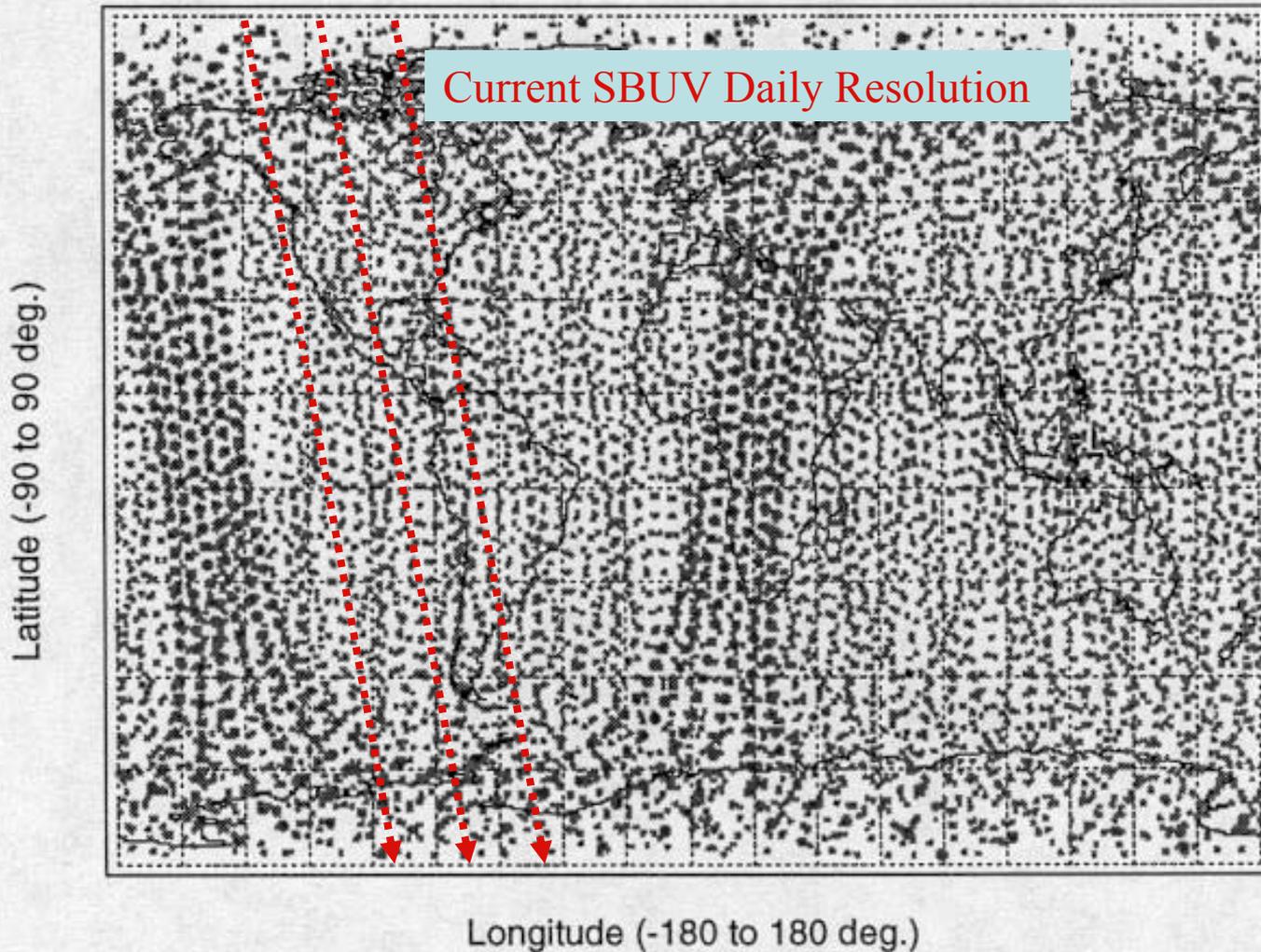
Latitude	70	75	80	85	90	95
26	41	41	41	41	41	41
24						
22	34	34	34	34	34	34
20						
18	32	33	33	33	33	33
16						

Latitude	70	75	80	85	90	95																
26	42	43	43	43	43	45	45	47	47	49	49	50	51	51	52	52	51	51	51	49	49	
24	42	43	42	42	43	44	44	45	46	48	48	49	49	50	50	50	50	49	48	46	49	
22	39	39	40	40	41	40	41	42	43	44	44	43	42	42	42	42	42	42	43	44	44	
20	36	37	38	39	39	39	40	41	42	42	43	43	42	41	40	40	39	38	38	38	39	40
18	33	34	36	36	36	36	37	38	40	42	42	41	40	39	39	39	39	39	40	42	44	43
16	32	32	34	36	35	36	36	36	36	38	39	38	38	38	38	37	37	38	38	39	40	41

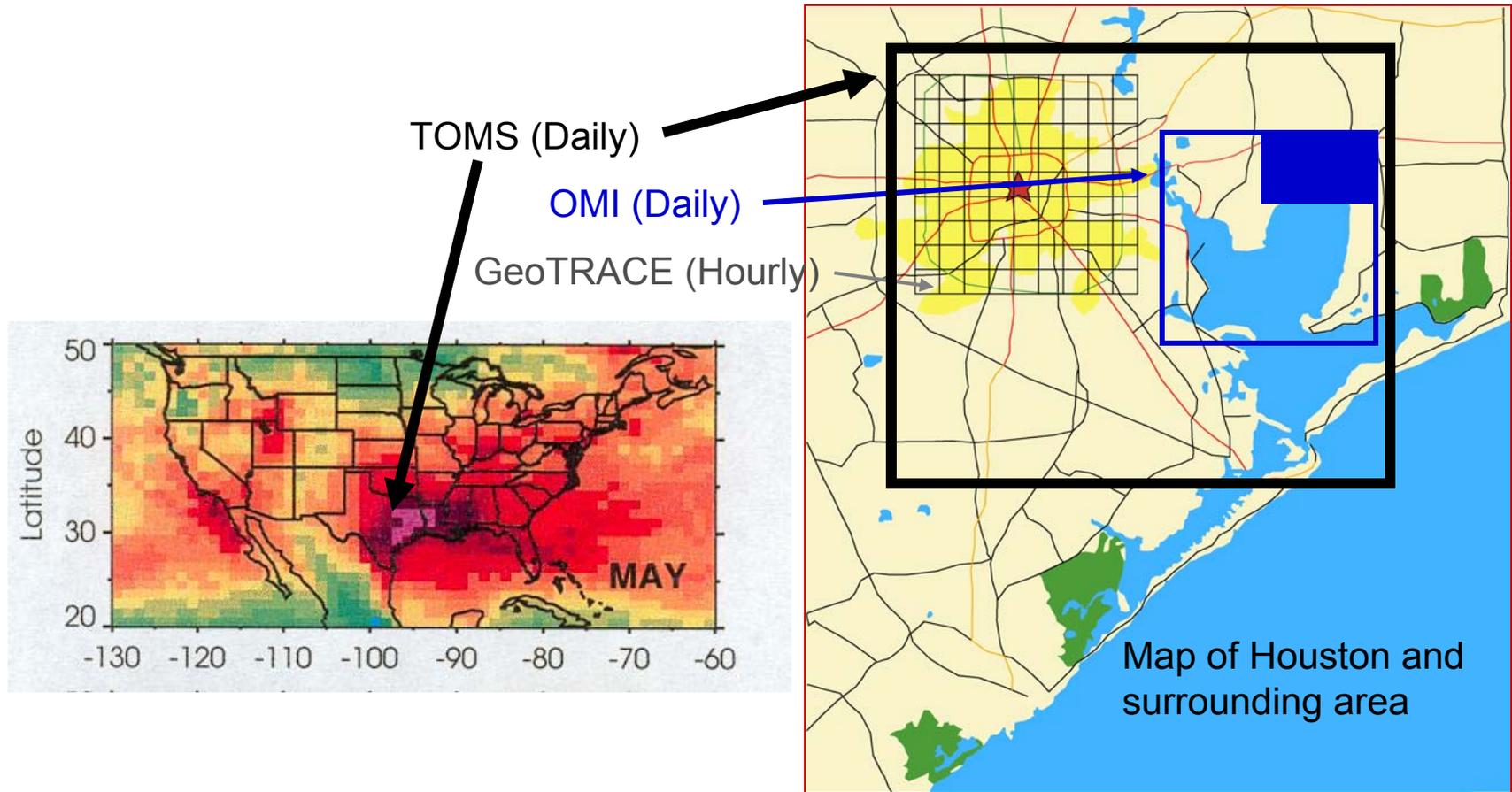
Future Satellite Measurements Will Improve Upon Techniques Developed with TOMS Measurements

- OMI and Other Aura Instruments Will Improve Data Resolution
- Data Assimilation and New “Smart” Interpolation Schemes Will Resolve Daily Stratospheric Ozone Distribution Better
- Eventual Goal is Geostationary TOMS-like Measurement Capability to Resolve Tropospheric Transport

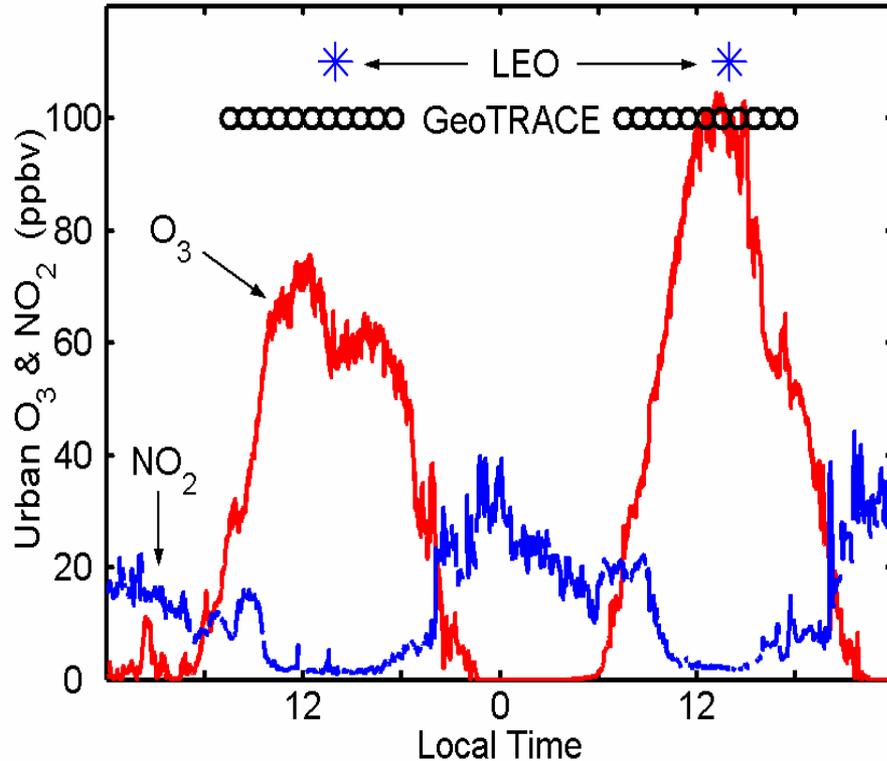
HIRDLS Daily Profile Coverage Will Provide Sufficient Information to Derive 3-Dimensional Stratospheric Ozone Distribution Down to 1 km Below Tropopause



Geostationary Observations Will Provide Hourly Observations with 5-km Resolution



Geostationary Observations Capture Diurnal Variability



O_3 , aerosols, & precursors change rapidly during the day.

Stars indicate typical times for LEO measurements.

Circles indicate individual GeoTRACE measurements.

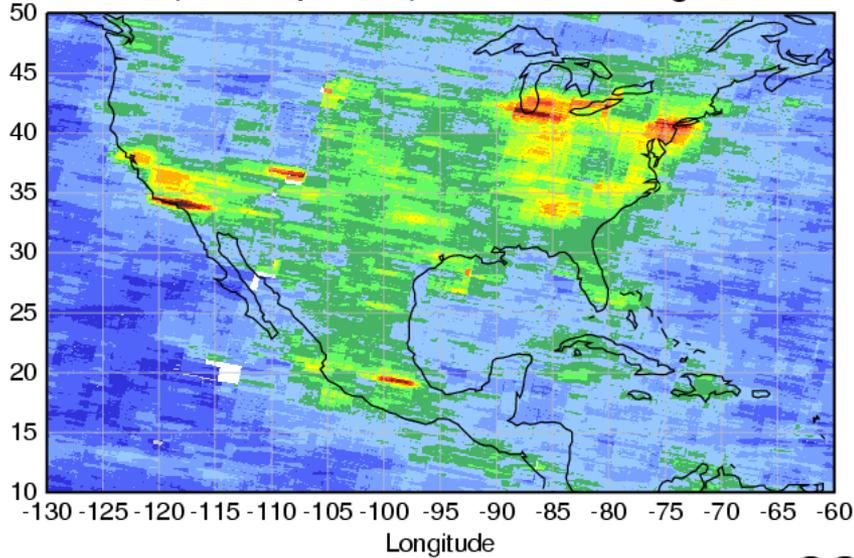
Summary

- TOMS Not Designed to Make Tropospheric Measurements
 - **But It DID!!!**
- Tribute to How Well It Worked and the Dedication of the Team Providing and Maintaining the Quality of the Data
- Not Perfect: But Provided Unimaginable Global Insight into How Regional Processes Affect Tropospheric Trace Gas Distributions
- Issues to Be Addressed
 - What is TOMS Sensitivity in Lower Atmosphere?
 - How Can the Stratosphere Be Removed from the Total Ozone Column?

BACK-UP SLIDES

Effects of spatial resolution: North America

GOME (SCIA pixels) NO₂ ex.: August 2002



Maximum values

GOME 7E15 molec/cm²

SCIA 17E15 molec/cm²

Match GOME and SCIA Pixels

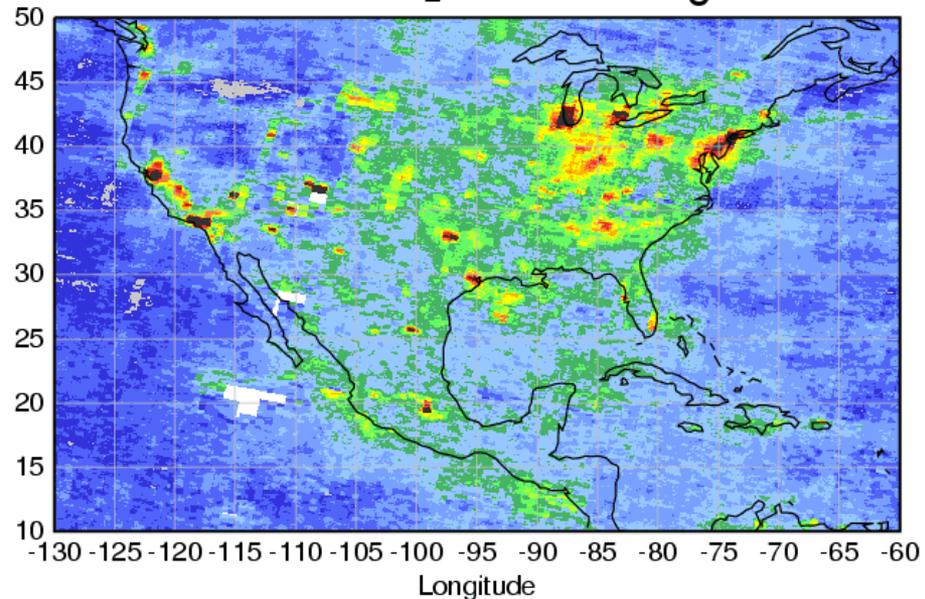
Resolution

GOME

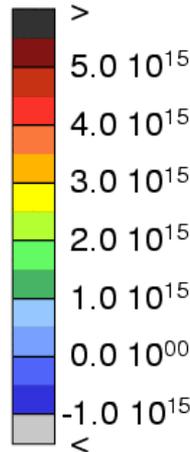
SCIA

OMI

SCIAMACHY NO₂ excess: August 2002

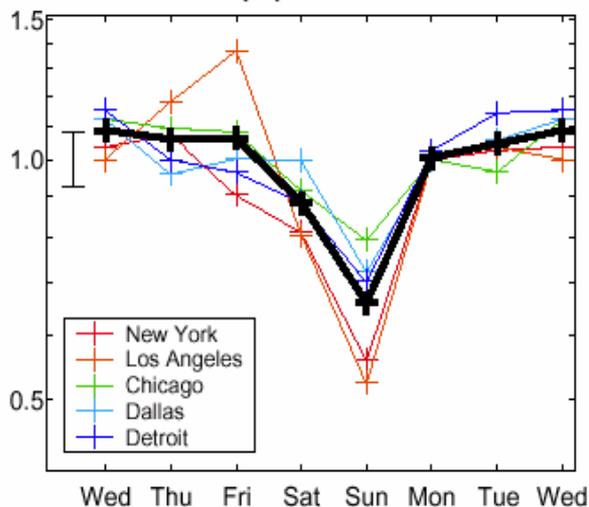


VC NO₂
[molec cm⁻²]



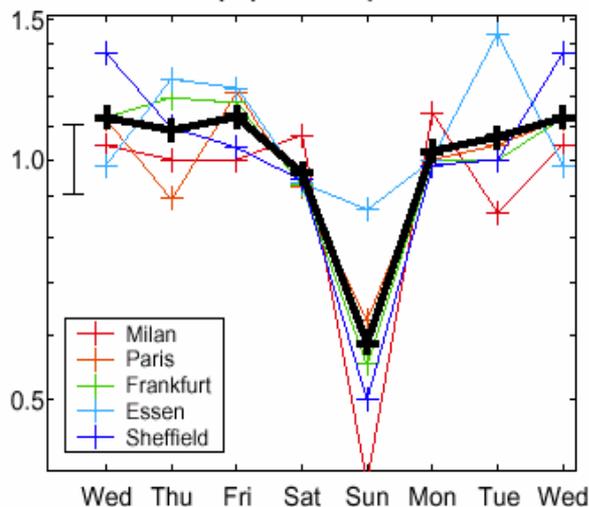
Time Resolution: Remote Sensing of the Sabbath

(1) USA



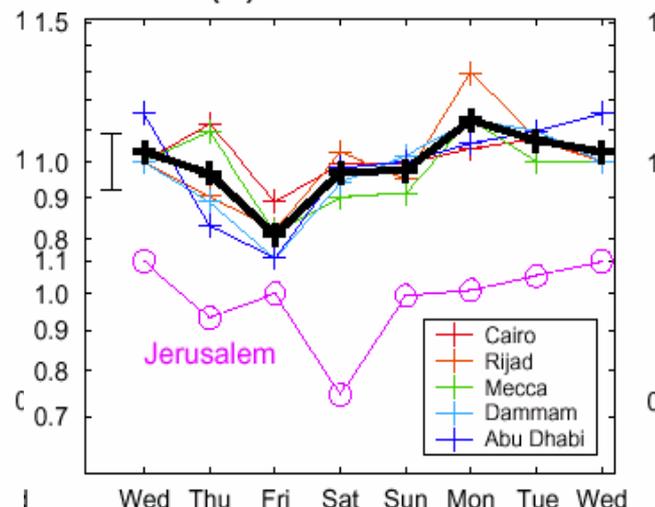
Sun

(2) Europe



Sun

(4) Middle East

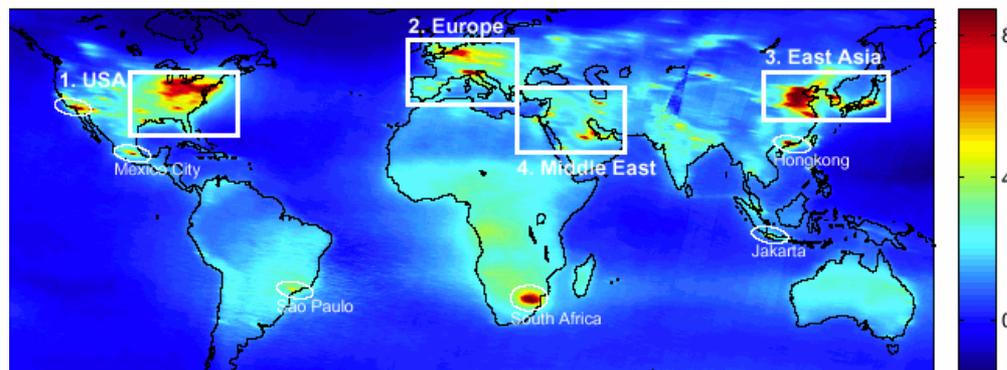


Fri

Sat

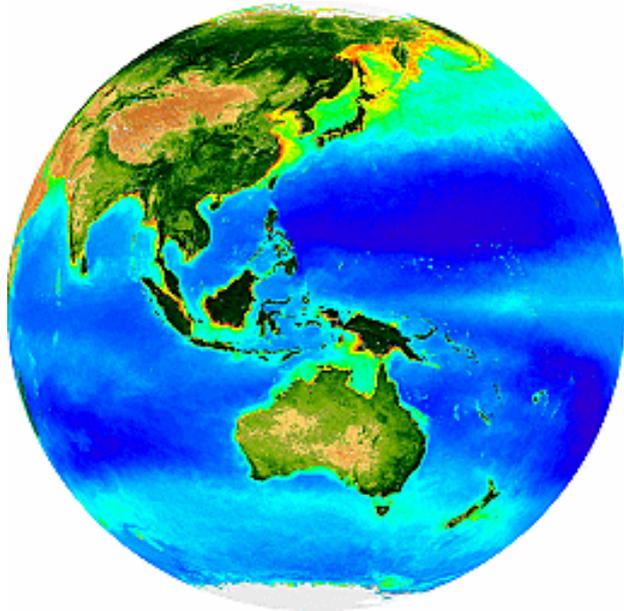
NO₂ is produced by combustion.

There is less combustion (energy production) on the “Days of Rest.”

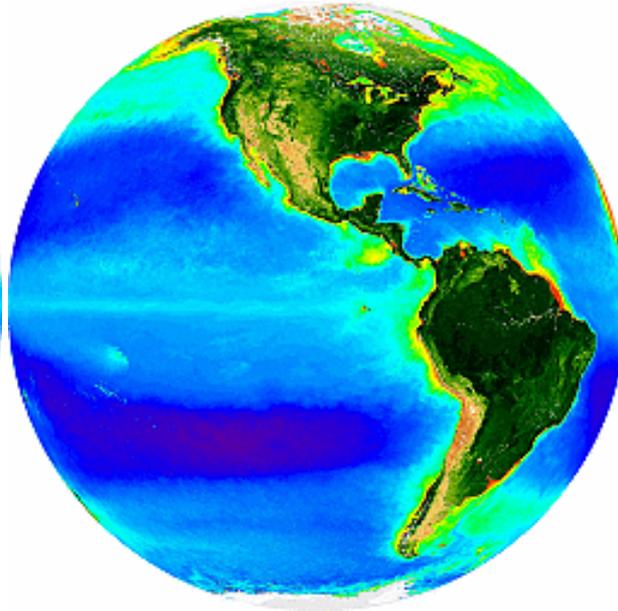


International Cooperation

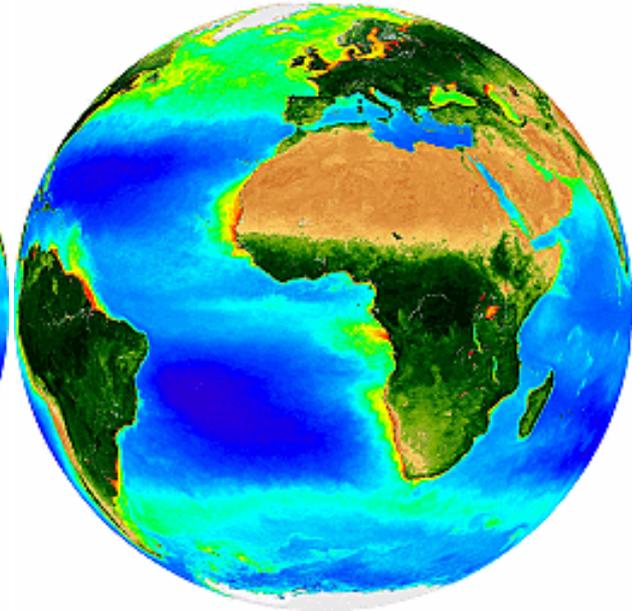
Complementary Coverage



NASDA



NASA

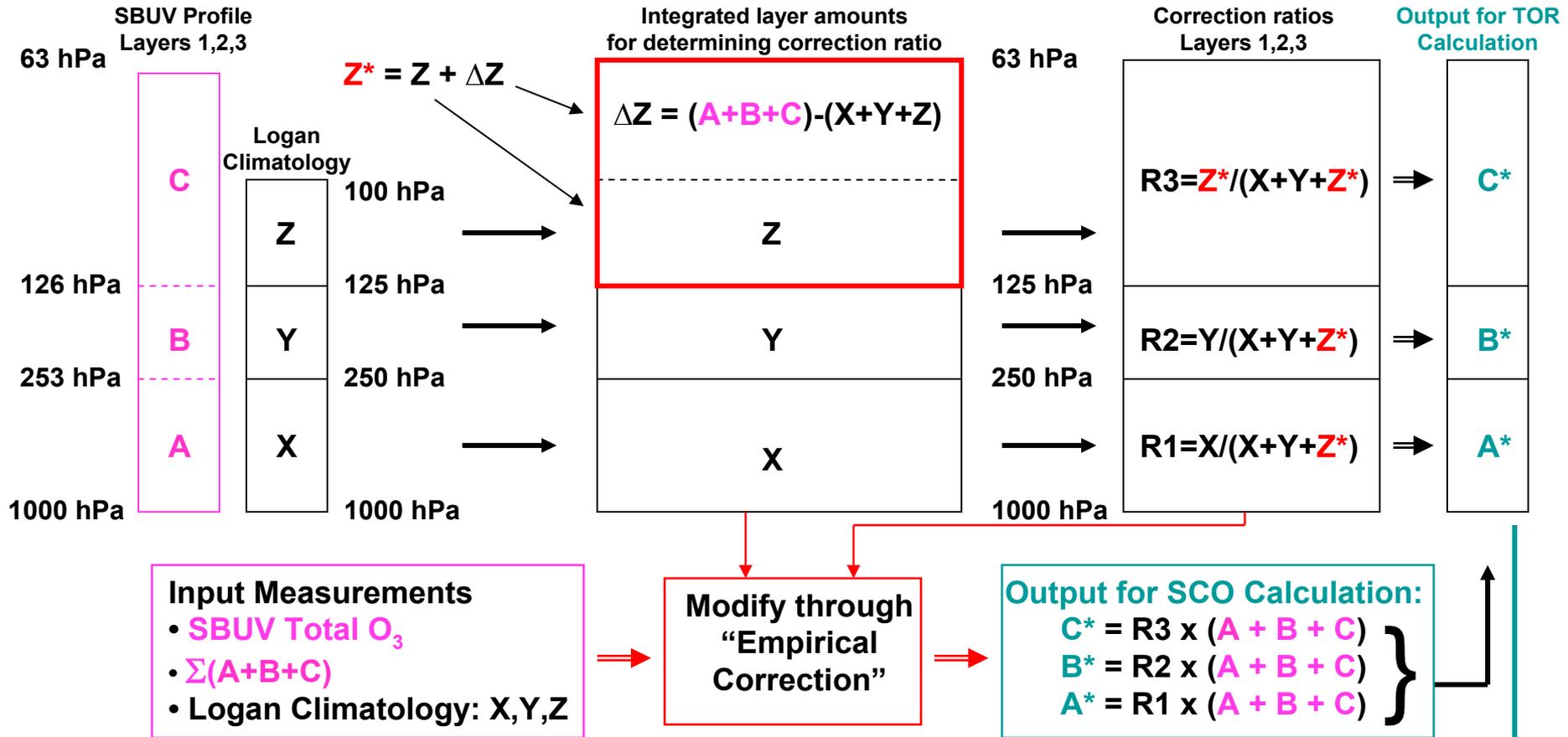


ESA

ESA has received geostationary proposals. NASDA is doing studies.

Calculation of TOMS/SBUV Tropospheric Ozone Residual

Part I: Calculate Stratospheric Column Ozone (SCO)



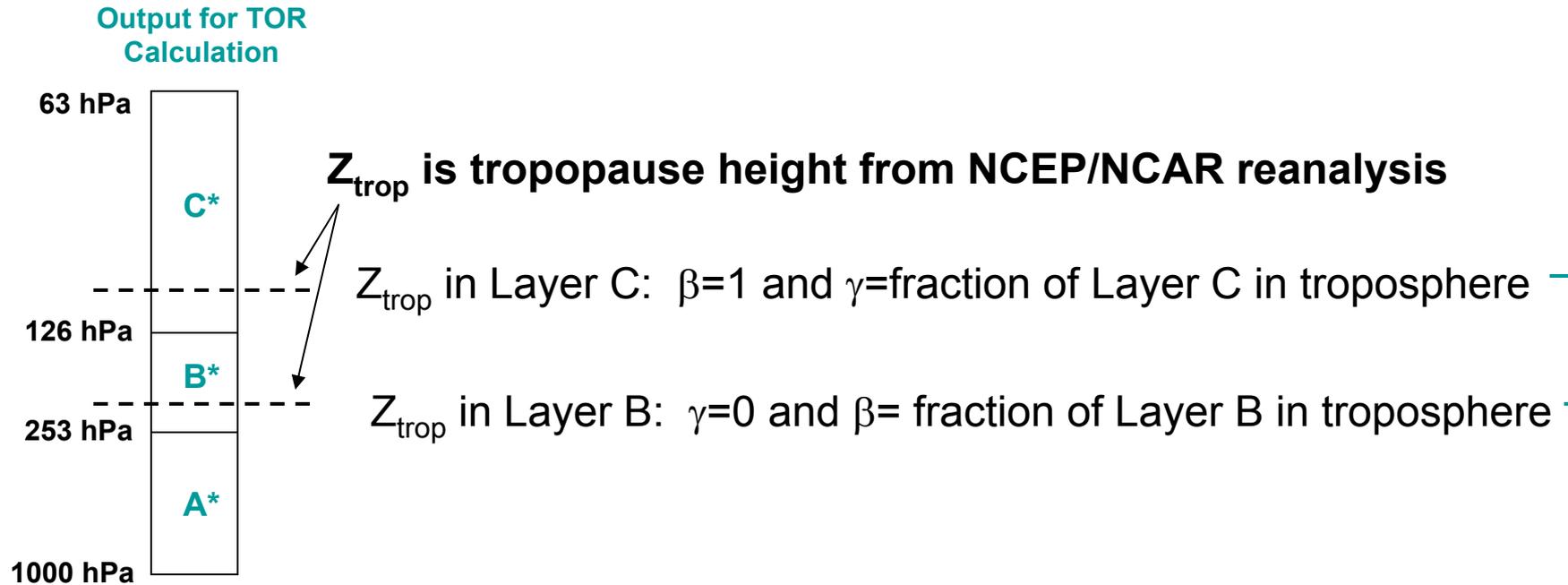
Part II: Calculate TOR from TOMS Total O_3 and SCO

$$(2a) \text{ SCO} = \text{SBUV Total } O_3 - \gamma C^* - \beta B^* - A^*$$

$$(2b) \text{ TOR} = \text{TOMS Total } O_3 - \text{SCO}$$

Note: γ and β are values between 0 and 1 and are determined by NCEP/NCAR Reanalysis tropopause height

Define fractional coefficients (β and γ) for TOR calculation



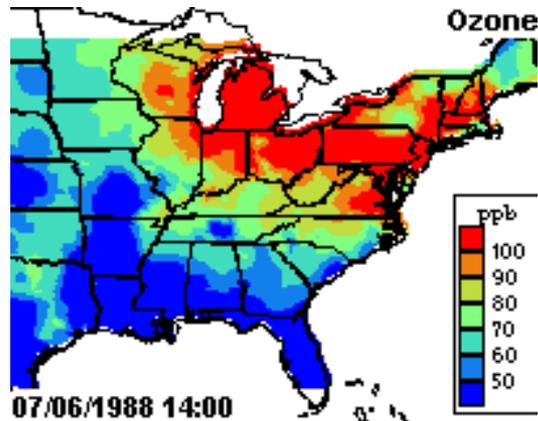
Part II: Calculate TOR from TOMS Total O₃ and SCO

$$(2a) \text{ SCO} = \text{SBUV Total O}_3 - \gamma C^* - \beta B^* - A^*$$

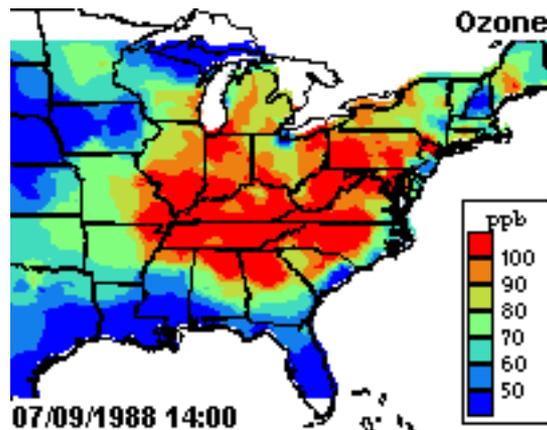
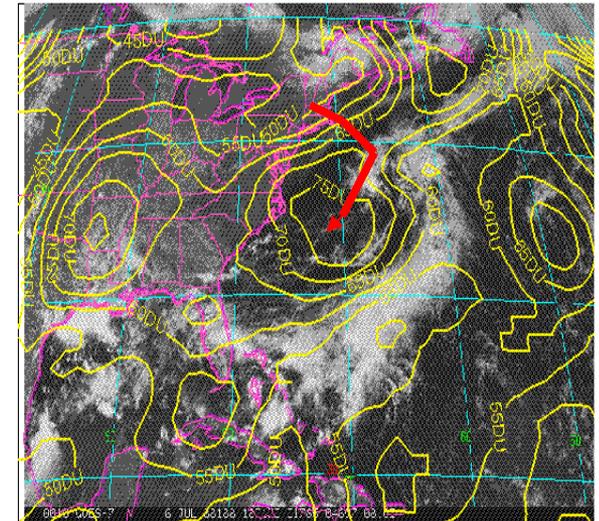
$$(2b) \text{ TOR} = \text{TOMS Total O}_3 - \text{SCO}$$

Note: γ and β are values between 0 and 1
if Z_{trop} is in Layer A, TOR is not calculated

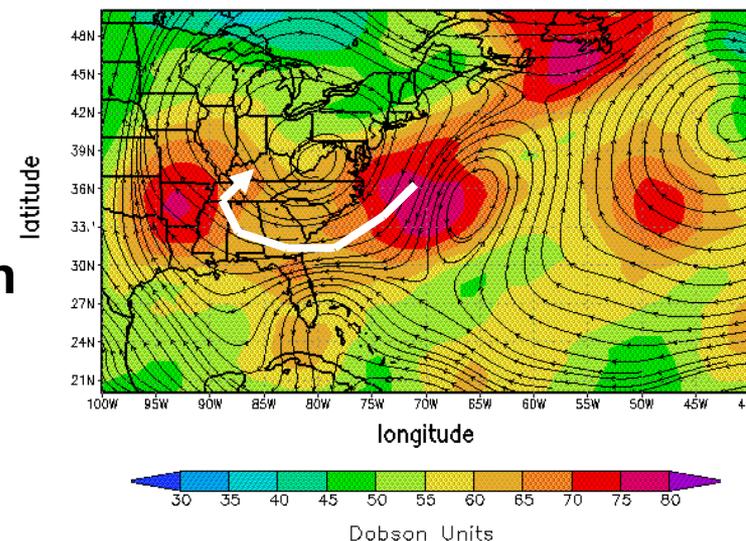
Satellite Study Demonstrates Synoptic-Scale Pollution Transport



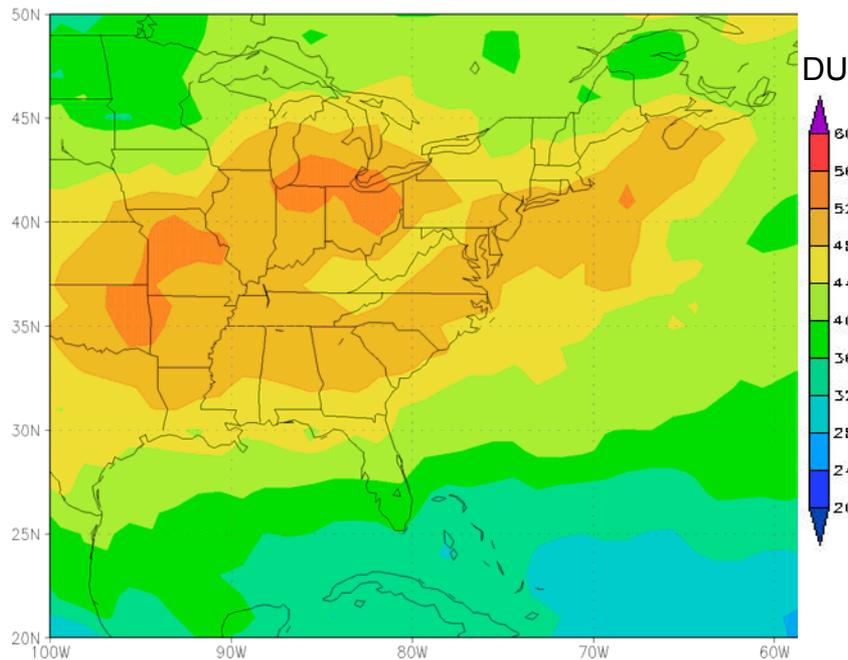
Pollution from northern states pools off North Carolina coast



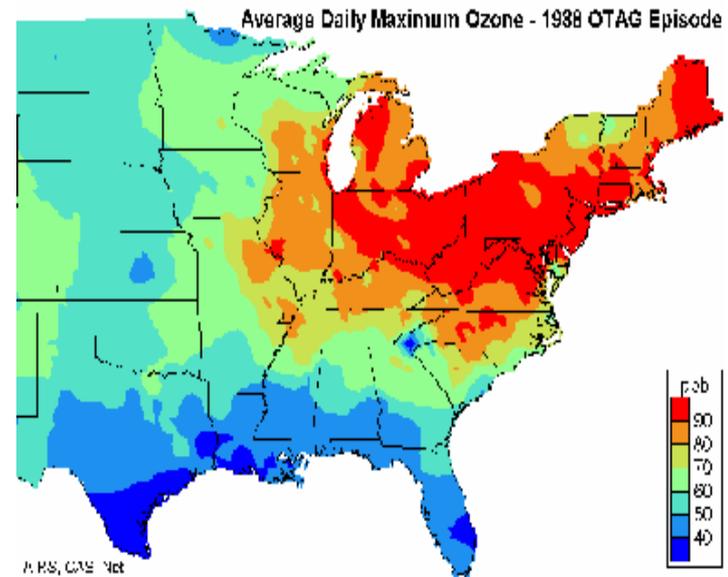
Unique transport situation carries off-shore pollution to southern states



July 1988 Monthly TOR Captures High Ozone During Major Pollution Episode

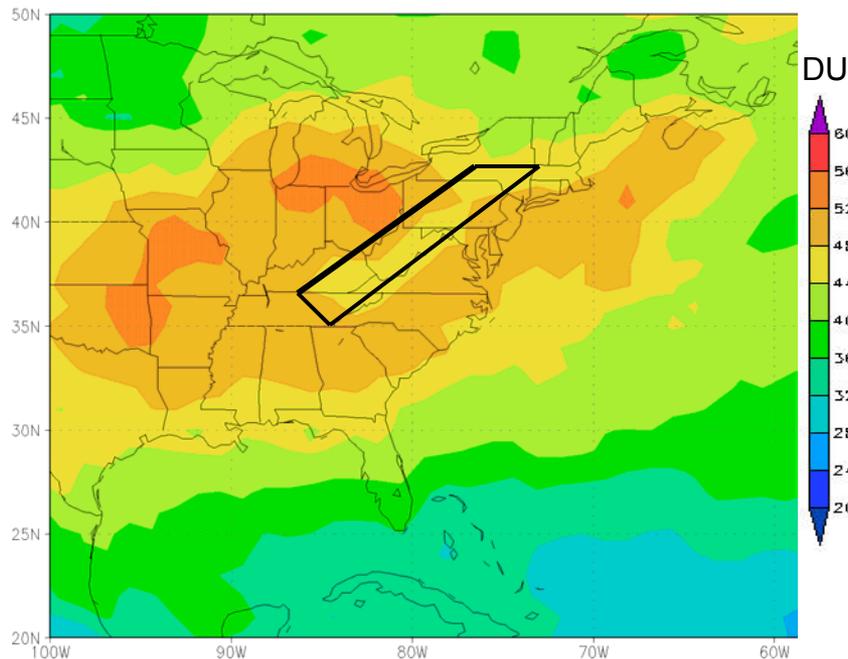


July 1988 TOR

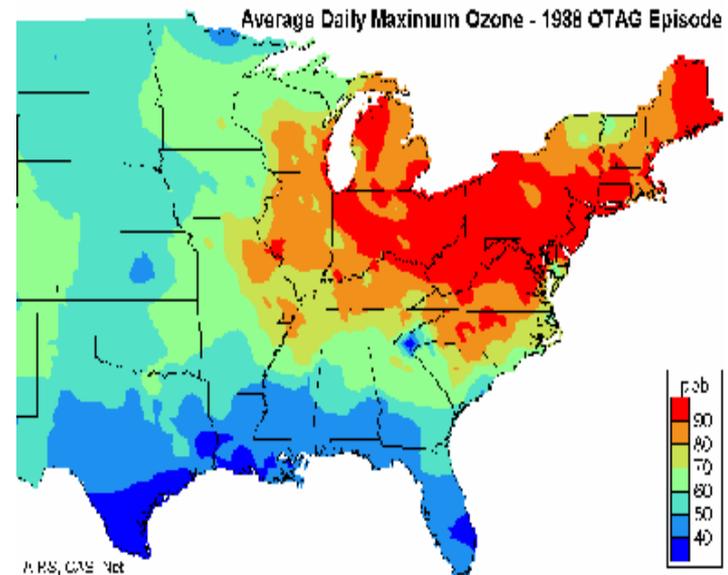


July 3-15 Average Daily Max. O_3
(from Schichtel and Husar, 1998)

July 1988 Monthly TOR Captures High Ozone During Major Pollution Episode



July 1988 TOR

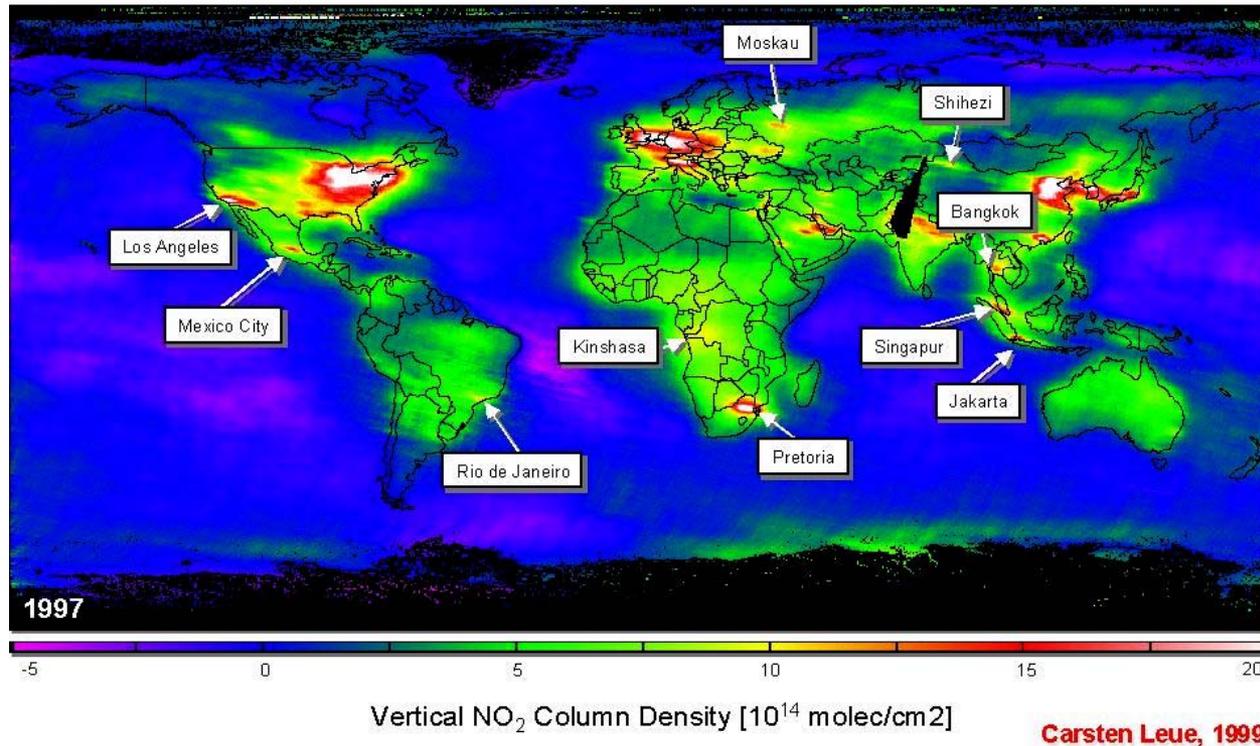


July 3-15 Average Daily Max. O_3
(from Schichtel and Husar, 1998)

- Lower TOR within box due to terrain artifact
- Use terrain information for global validation

GOME NO₂ Measurements Also See Enhancements over India and China

Average Tropospheric NO₂ Column Density During 1997, GOME

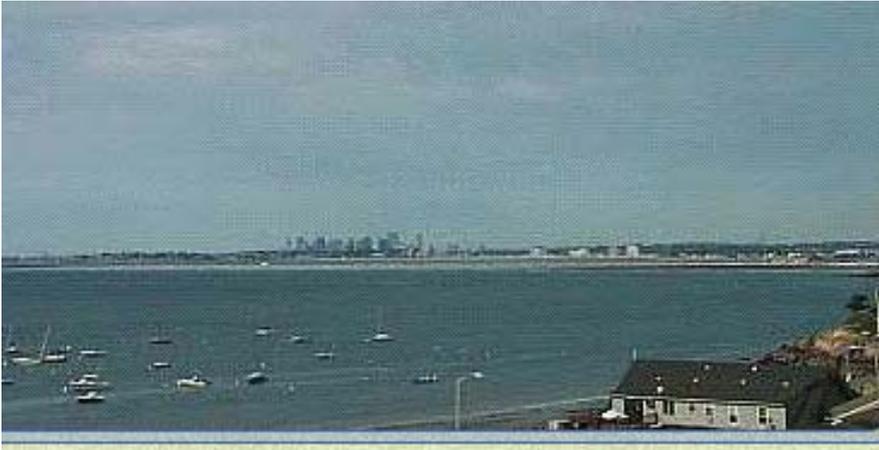


Global TOR Averages Change with TOMS Archive

- Fishman et al. [1990]: **32.7 DU** (pseudo-Version 6/SAGE)
Version 6 corrected for instrument drift
- Fishman & Brackett [1997]: **27.5 DU** (Version 7/SAGE)
Version 7 incorporates ISCCP cloud climatology for correction
- Fishman et al [2003]: **31.5 DU** (pseudo-Version 8/SBUV)
Version 8 includes aerosol and scan-angle dependence corrections

Why do we need time resolution?

Boston Morning



Boston Afternoon



Emissions and Ozone/Aerosol production change.

Air Quality changes during the day.